MASTER COPY KEEP THIS COPY FOR REPRODUCTION PURPOSES

REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY *(Leave blank)*

2. REPORT DATE
11/08/94

3. REPORT TYPE AND DATES COVERED
Final Report: 8/12/92-6/30/94

4. TITLE AND SUBTITLE
Growth, Characterization and Application of KTN Family Nonlinear Optical Crystals

5. FUNDING NUMBERS
DAAL03-91-G-0305

6. AUTHOR(S)
Amnon Yariv, Anthony Kewitsch, Rudy Hofmeister

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
Department of Applied Physics, 128-95
California Institute of Technology
Pasadena, CA 91125

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)
U. S. Army Research Office
P. O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING / MONITORING AGENCY REPORT NUMBER
ARO 29321.3-PH

11. SUPPLEMENTARY NOTES
The view, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

12a. DISTRIBUTION / AVAILABILITY STATEMENT

Approved for public release; distribution unlimited.

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

We have recently grown KLTN:(Cu, Ni, Ni/Ti) and SBT crystals and characterized their nonlinear-optical properties. The growth of these high quality crystals is a result of recent improvements in the crystal growth system; namely, improved flux uniformity and higher operating temperature. These and other materials have been characterized for application to holographic data storage and quasi-phase matched second harmonic generation. In particular, the paraelectric KLTN materials doped with Ni exhibit an enhanced long wavelength sensitivity (> 600 nm), which is desirable for recording holograms with ubiquitous He-Ne or semiconductor lasers. Furthermore, we have discovered that dynamic ferroelectric domain gratings in addition to the more common electrooptic gratings are present in SBN, a close relative of SBT. This has important applications to permanent fixing of volume holograms and tunable quasi-phase matching. To complement this experimental work, we have also derived analytical expressions for the two beam coupling process which allows us to deduce photorefractive material parameters for these crystals. This balance of experimental and theoretical work has enabled us to continually develop new materials whose properties are optimized for nonlinear optical applications.

14. SUBJECT TERMS
Photorefractive Crystals, Coupled Beams, Holograms

15. NUMBER OF PAGES
6

16. PRICE CODE

DTIC QUALITY INSPECTED 4

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

NSN 7540-01-280-5500

Standard Form 298 (Rev. 2-89)
Prescribed by ANSI Std. Z39-18
298-102

Highlights of Research Program
ARO/DARPA DAAL03-91-G-0305

Growth, Characterization and Application of KTN Family Nonlinear Optical Crystals

Amnon Yariv	Principal Investigator
Anthony Kewitsch Doruk Engin Xiaolin Tong	Graduate Students
Ron Agranat	Visiting Associate
Rudy Hofmeister Moti Segev	Research Associates

BRIEF OUTLINE OF RESEARCH FINDINGS:

1. Photorefractive Crystal Growth

Significant progress was made in photorefractive crystal growth during the course of this research.

The successful growth of KLTN materials doped with copper was achieved. In addition, the crystal growth process was improved to allow growth of crystals with maximum photorefractive response near room temperature. This result was achieved by adjusting the composition of the crystal to a higher concentration of niobium while retaining good optical quality.

The growth of KLTNs doped with nickel and with a nickel/titanium combination were also demonstrated. Investigation of these materials' photorefractive properties has yielded interesting results. These materials have been shown to respond much more strongly to longer wavelength light than their copper -doped counterparts. Two-beam diffraction efficiencies of well over 65% are now possible in 2-3mm thick crystals using red light at 633nm.

We have successfully implemented a new crystal growth system and have grown several dozen high-quality crystals[1]. The new system incorporates numerous improvements over the previous system; these include a better flux mixing

capability to give more uniform crystal compositions. In addition, the new system can operate at a higher temperature - up to 1650°C.

Research in the development of strontium barium titanate (SBT) is continuing with the new crystal growth system. The material growth parameters were initially mapped out, such as the phase diagram of the system and we then performed several preliminary growths. The next effort was to generate a seed material from which to grow larger crystals. This step has been accomplished and the research is currently proceeding to larger crystal growth development.

At present, the crystal growth effort is continuing with the objective of improving the photorefractive wavelength response of the materials. Additionally, we plan to produce high quality SBT material and characterize its material properties.

References

[1] R. Hofmeister, A. Yariv, and A. Agranat, "Growth and characterization of the perovskite KLTN:Cu," J. Crystal Growth 131 486 (1993).

2. Analysis of Coupled Beams in Photorefractive Media

We developed a new method of solving coupled differential equations. The particular equations solved are the type typically encountered in describing grating formation in photorefractive materials. The recently developed methods were applied to two particular problems. First, they were used to explain experimental results of the frequency response of fixed photorefractive interference filters[1]. These results agreed favorably with experimental data supplied by Accuwave corporation of their solar filter. Second, they have allowed a theoretical solution of the diffraction results seen in holographic gratings written in paraelectric KLTN[2]. Again, the theory agreed well with the data. The equations were also used to develop a new technique for determining the photorefractive phase (a material parameter). These results are now used as a general tool in the solution of these types of equations.

References:

[1] R. Hofmeister, A. Yariv, and S. Yagi, "Spectral Response of Fixed Photorefractive Grating Interference Filters," JOSA B.

[2] R. Hofmeister and A. Yariv, "Diffraction and beam coupling in paraelectric KLTN," in preparation.

3. Progress in Fixing of Photorefractive Holograms

In the last 2 years we have succeeded in generating dynamic ferroelectric domain gratings using only photoinduced space charge fields in $Sr_{.75}Ba_{.25}Nb_2O_6$ (SBN:75). We have demonstrated that these initially dynamic domain gratings can be fixed by a variety of techniques. For instance, domain gratings are permanently fixed after writing with high intensity beams, if the exposure is above 100 J cm^{-2}. Recently, we have reduced this threshold by two orders of magnitude by using a short exposure technique in which the permanent domain grating is formed by developing the grating in the dark following an initial exposure. In support of the early fixing studies in SBN, we have found that the diffraction efficiency of the remnant domain gratings can enhanced by applying a depoling field pulse in the dark; however, domain gratings may be fixed without any external field other than the photogenerated space charge field. We have conclusively determined that the remnant gratings are indeed ferroelectric domain gratings and have theoretically and experimentally studied the implications of SBN's glassy polarization phase on the domain grating formation.

We have used this technique to permanently store volume holograms in SBN for data storage applications. A unique advantage of this technique is the ability to selectively overwrite existing fixed holograms at predetermined addresses. This overcomes the primary drawback of the ion drift fixing mechanism in $LiNbO_3$; that is, the inability to readily update the memory following fixing. On the other hand, the dynamic space charge holograms, while being updatable, are destroyed upon readout. Thus, this selective, page addressable fixing technique has several potential applications in a high capacity, high data rate read/write holographic optical memory with long term storage capabilities.

Periodic domain gratings can also be used to achieve quasi-phase matched (QPM) second harmonic generation. By writing an optical grating with a period equal to twice the coherence length of the interaction, the second harmonic conversion efficiency can be significantly increased above the background level. Several domain gratings can be written in the same volume to tailor the spectral response of the QPM peak. Because these domain gratings are dynamic, the grating period can updated on very short time scales (< seconds) to enable tunable QPM. To render the domain gratings permanent, they can be frozen-in using the fixing techniques developed for the permanent storage of volume holograms.

LIST OF MANUSCRIPTS SUBMITTED OR PUBLISHED UNDER ARO SPONSORSHIP DURING THIS REPORTING PERIOD, INCLUDING JOURNAL REFERENCES:

- A. Agranat, R. Hofmeister, and A. Yariv, "Characterization of a new photorefractive material: $K_{1-y}L_yT_{1-x}N_x$," Optics Letters 17, 10. p. 713 (May 1992).

- R. Hofmeister, A. Yariv, S. Yagi and A. Agranat, "New photorefractive mechanism in centrosymmetric crystals: a strain-coordinated Jahn-Teller," submitted for publication to Physical Review Letters.

- R. Hofmeister and A. Yariv, "Vibration detection using dynamic photorefractive gratings in KTN/KLTN crystals," submitted for publication to Applied Physics Letters.

- R. Hofmeister, A. Yariv, and S. Yagi, "Spectral response of fixed photorefractive grating interference filters," to appear JOSA B.

- R. Hofmeister, A. Yariv, A. Agranat, "Growth and characterization of the perovskite $K_{1-y}Li_yTa_{1-x}Nb_xO_3$:Cu, Journal of Crystal Growth, vol. 131, pp. 486-494 (1993).

- R. Hofmeister, A. Yariv, A. Kewitsch and S. Yagi, "Simple methods of measuring the net photorefractive phase shift and coupling constant," Optics Letters, vol. 18, no. 7, pp. 488-490 (April 1993).

- A. Kewitsch, M. Segev, A. Yariv, "Permanently Fixed Volume Phase Gratings in Ferroelectrics," Chapter 9, The Photorefractive Effect, Ed. D. Nolte, Kluwer Academic (1994).

- A. Kewitsch, A. Saito, M. Segev, A. Yariv, R. Neurgaonkar, "Optical and electrical barkhausen noise induced while recording ferroelectric domain holograms," submitted to JOSA B (July 1994).

- A. Kewitsch, M. Segev, A. Yariv, G. Salamo, T. Towe, E. Sharp, R. Neurgaonkar, "Ferroelectric domain gratings induced by photorefractive space charge fields," Phys. Rev. Lett. 73 (1994).

- A. Kewitsch, M. Segev, A. Yariv, G. Salamo, T. Towe, E. Sharp, R. Neurgaonkar, "Tunable quasi-phase matching using dynamic ferroelectric domain gratings induced by photorefractive space-charge fields," Appl. Phys. Lett. 64, 3068 (1994).

- A. Kewitsch. M. Segev, A. Yariv, R. Neurgaonkar, "Controlled, periodic domain formation in sbn induced by volume holograms," Jpn. J. Appl. Phys. 32, 5445 (1993).

- A. Kewitsch, M. Segev, A. Yariv, R. Neurgaonkar, "Selective, page-addressable fixing of volume holograms in strontium barium niobate," Opt. Lett. 18, 1262 (1993).

- A. Kewitsch, M. Segev, A. Yariv, R. Neurgaonkar, "Electric-field multiplexing/demultiplexing of volume holograms in photorefractive media," Opt. Lett. 18, 534 (1993).

REPORT OF INVENTIONS:

- An all-optical, self aligning holographic vibration sensor utilizing the zero external electric field photorefractive effect in strained paraelectric materials. U.S. Patent granted

- Method of high quality growth of transition-metal doped $K_{1-y}L_yT_{1-x}N_x$ crystals that exhibit the zero external field photorefractive effect. Patent applied for.

- Major, systematic characterization of $K_{1-y}L_yT_{1-x}N_x$, photorefractive crystals[1]. Major improvement in optical quality of KTN nonlinear crystals.

- Discovered new photorefractive mechanism, based on Jahn-Teller relaxation in KTN[2].

- Design and fabrication of improved crystal growth furnace system, suitable for use at up 1650°C